Critical Fluctuations in the Microwave Complex Conductivity of BSCCO and YBCO Thin Films

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Critical fluctuations above T_c are studied in the microwave complex conductivity of BSCCO-2212, BSCCO-2223, and YBCO thin films. The analysis of the experimental data yields the absolute values and the temperature dependence of the reduced coherence length $\xi(T)/\xi_0$. Besides the well known *3D* XY critical regime having the static critical exponent $\nu = 2/3$, a crossover to a new critical regime with $\nu = 1$ is observed when T_c is approached. In more anisotropic superconductors the reduced coherence lengths are larger, and the critical regimes extend to higher temperatures.

The nature of superconducting fluctuations in high-temperature superconductors has continued to attract a great deal of attention. The small coherence lengths, high transition temperatures, and large anisotropy due to the layered structure make the fluctuations of the order parameter much stronger than in classical low temperature superconductors. Particularly intriguing has appeared the possibility of observing critical fluctuations in a fairly wide temperature range around T_c. In this regime the critical exponents deviate with respect to their mean-field values. Renormalization group theory provides an appropriate description of this phenomenon. One obtains scaling laws and universality features which are of great importance in identifying the nature of the phase transition [1]. The critical fluctuations in high-T_c superconductors have been studied by penetration depth [2, 3], specific heat [4, 5], magnetization [4], resistivity [6, 7, 8], thermal expansivity [9, 10], and two-coil inductive measurements [11]. The conclusions drawn from these data seemed to point at the critical behavior pertaining to the *3D* XY universality class in high-T_c superconductors. However, the discrepancies in the region of critical behavior varied from a fraction of a degree [7, 8] to ± 10 K [2, 9].

In this paper we present novel results and a new type of data analysis of the microwave fluctuation conductivity in thin films of high temperature superconductors $Bi_2Sr_2CaCu_2O_{8+\delta}$ (BSCCO-2212), $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ (BSCCO-2223), and $YBa_2Cu_3O_{7-\delta}$ (YBCO). The advantage of the *ac* method is that one obtains two experimental data sets, i. e. the real and imaginary parts of the fluctuation conductivity, both of which have to corroborate with a given theoretical model using the same set of parameters. It represents therefore a more stringent test on the nature of the fluctuations than methods yielding a single experimental curve. Using this feature, we determine for the first time experimentally the absolute values and the temperature dependence of the coherence length above T_c in high-Tc superconductors. Quite surprisingly, we observe in all our samples two critical regimes with the static critical exponents $\nu = 2/3$ well above T_c, and a crossover to $\nu = 1$ when T_c is approached.

YBCO thin films having 200 nm thickness were grown on MgO substrate. BSCCO-2212 (350 nm) and BSCCO-2223 (100 nm) thin films were grown on $LaAlO_3$ and $NdGaO_3$ substrates, respectively. The sample was positioned in the center of an elliptical copper cavity resonating in $_e\text{TE}_{111}$ mode at ≈ 9.5 GHz, with the microwave electric field E_ω parallel to the *ab*-plane of the superconducting film. The temperature of the sample could be varied from 2 K to room temperature by a heater and sensor assembly mounted on the sapphire sample holder. The Q-factor was measured by a recently introduced modulation technique [12], which enables the resolution of $\Delta(1/2Q)$ to 0.02 ppm, while the frequency shift was monitored by a microwave frequency counter. Fig. 1 shows the experimental values of $\Delta(1/2Q)$ and $\Delta f/f$ for the BSCCO-2212 thin film. The complex frequency shift $\Delta\widetilde{\omega}/\omega = \Delta f/f + i\,\Delta(1/2Q)$ is related to the sample and cavity parameters according to the cavity perturbation analysis [13]

$$\frac{\Delta\widetilde{\omega}}{\omega} = \frac{\Gamma}{N}\left[1 - N + \frac{(\widetilde{k}/k_0)^2 N}{\left[\coth(i\widetilde{k}d/2) + \tanh(i\widetilde{k}\zeta)\right]i\widetilde{k}d/2}\right]^{-1}, \tag{1}$$

where Γ is the filling factor of the sample in the cavity, and N is the depolarization factor of the film. The complex wavevector in the film is $\widetilde{k} = k_0\sqrt{1 - i\widetilde{\sigma}/(\epsilon_0\omega)}$, where $k_0 = \omega\sqrt{\mu_0\epsilon_0}$ is the vacuum wavevector, and $\widetilde{\sigma} = \sigma_1 - i\sigma_2$ is the complex conductivity of the film. The thickness of the film is d, and ζ is the asymmetry parameter due to the substrate [13]. The parameters N and Γ in Eq. (1) have been evaluated from the comparison of the experimental and
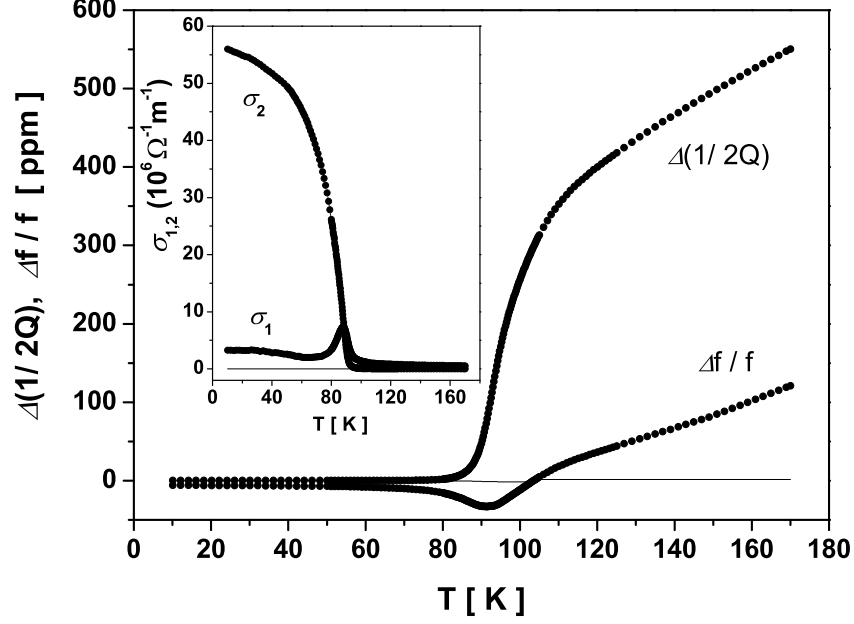
FIG. 1: Temperature dependences of the experimentally measured $\Delta(1/2Q)$ and $\Delta f/f$ in a BSCCO-2212 thin film. The inset shows the real (σ_1) and imaginary (σ_2) parts of the complex conductivity.

theoretical curves of $\Delta(1/2Q)$ and $\Delta f/f$ in the normal state far above T_c where σ_n is known from dc measurements. By taking the ratio of the slopes of the theoretical $\Delta(1/2Q)$ and $\Delta f/f$ curves, one eliminates Γ so that N can be determined from the comparison to the experimental ratio. In the next step, Γ can be determined from the measured value of $\Delta(1/2Q)$ at a given temperature in the normal state. We have used $\sigma_n = 6.4 \cdot 10^5 \ \Omega^{-1}m^{-1}$ for the normal state conductivity at 150 K. Similar evaluation at temperatures above 150 K did not change the resulting parameters. Eq. (1) can then be used to convert the experimental data for $\Delta(1/2Q)$ and $\Delta f/f$ at any temperature to obtain the corresponding experimental values of σ_1 and σ_2 as shown in the inset to Fig. 1. Similar results for the complex conductivity in BSCCO-2212 have been obtained previously on single crystals [14]. The observed peak in σ_1 is due to the superconducting fluctuations and its maximum is reached when the coherence length diverges [1, 15]. We found $T_c = 87.9$ K in our BSCCO-2212 thin film.

In this paper we focus on σ_1 and σ_2 at, and above T_c, shown on an expanded scale in Fig. 2(a). Here the reduced temperature $\epsilon = \ln(T/T_c)$ is used. The ac fluctuation conductivity above T_c is given by [16]

$$\widetilde{\sigma}^{\,3D} = \frac{e^2}{32\hbar\xi_{0c}} \left(\frac{\xi(T)}{\xi_0}\right) \int_0^{Q_{ab}} \int_{-Q_c}^{Q_c} \frac{4\,q_{ab}^3 \left[1 - i\Omega(1 + q_{ab}^2 + q_c^2)^{-1}\right]}{\pi(1 + q_{ab}^2 + q_c^2)[\Omega^2 + (1 + q_{ab}^2 + q_c^2)^2]} \, dq_{ab}\, dq_c \ . \tag{2}$$

The prefactor is the Aslamazov-Larkin term for the $3D$ case. The reduced coherence length $\xi(T)/\xi_0$ is the same for the in-plane and c-axis coherence lengths, i. e. $(\xi_{ab}(T)/\xi_{0ab}) = (\xi_c(T)/\xi_{0c})$, so that we use it without subscripts. The cutoff in the fluctuation wavevector is introduced in $Q_{ab}(T) = k_{ab}^{max}\xi_{ab}(T) = \sqrt{2}\Lambda_{ab}\,\xi_{ab}(T)/\xi_{0ab}$ for the ab-plane and $Q_c(T) = k_c^{max}\xi_c(T) = \Lambda_c\,\xi_c(T)/\xi_{0c}$ along the c-axis, rather than a single cutoff on the modulus [17]. The dimensionless parameter $\Omega = (\pi\,\hbar\,\omega/16\,k_B\,T_c)\,(\xi(T)/\xi_0)^2$ involves the operating microwave frequency ω and the temperature dependence of the reduced coherence length. The analytical result of the integration in Eq. (2) and its application in the data analysis has been reported in detail [16]. The cutoff parameters Λ_{ab} and Λ_c are constrained by the ratio $\sigma_2(T_c)/\sigma_1(T_c)$ which can be evaluated from the experimental data. In the present case we found $\Lambda_{ab} = 0.7$ and $\Lambda_c = 0.06$. Also, the parameter ξ_{0c} in Eq. (2) can be determined from the experimental value of $\sigma_2(T_c)$, and we obtained $\xi_{0c} = 0.05$ nm for the BSCCO-2212 sample. Similar results for ξ_{0c} were obtained also from dc conductivity measurements [18].

The temperature dependence in the ac fluctuation conductivity of Eq. (2) is due to the reduced coherence length. It is interesting to examine first the simple Gaussian form $(\xi(T)/\xi_0) = 1/\sqrt{\epsilon}$. The fluctuation conductivity calculated

using the Gaussian coherence length is shown by the dotted lines in Fig. 2(a). Since the experimental values are much higher one may conclude that the fluctuations are not Gaussian in this temperature range. We have tried also other forms of the type $(\xi(T)/\xi_0) = A/\epsilon^\nu$, but none of those could fit satisfactorily the experimental data. Some examples are shown in the insets of Fig. 2. With the variation of the parameters A and ν, one can fit better either the section close to T_c, or the one at higher temperatures, or make a compromise which never gets the right shape in the whole temperature region. Obviously, a single static critical exponent ν cannot describe the experimental results. The true behavior of $\xi(T)/\xi_0$ has to be determined from the experimental data itself. Therefore, we have used the experimental values of $\sigma_2(T)$ to solve numerically for $\xi(T)/\xi_0$ in the imaginary part of $\widetilde{\sigma}^{\,3D}$ given by Eq. (2). The results are shown in Fig. 2(b). One observes that in the region $0.07 < \epsilon < 0.35$ the static critical exponent is close to $\nu = 2/3$ as expected for the critical regime of the $3D$ XY universality class. Thus, the present analysis recovers, in this part, the results obtained previously by other techniques in high-T_c superconductors. The solution of Eq. (2) it seems to yield correct results when used in the critical regime. Namely, the recent calculation by Wickham et al. [19] has shown that the form of the expression for the ac fluctuation conductivity does not depend on the regime of the fluctuations. Only the coherence length has different behavior in the Gaussian and critical regimes. In the present case, the full result in Fig. 2(b) shows a clear crossover from the $3D$ XY critical regime to another critical regime with $\nu = 1$ when T_c is approached. The appearance of these two critical regimes with a well defined crossover has not been reported previously. The absolute values of the reduced coherence lenght reveal that the crossover between the regimes is not just a change of the critical exponent but also involves a step in the magnitude of the coherence length.

On the high temperature side, one observes in Fig. 2(b) a deviation from the $3D$ XY critical regime at $\epsilon > 0.35$, probably marking the crossover to the Gaussian regime with the critical exponent $\nu = 1/2$. The drop seen in the last few points at $\epsilon > 0.5$ can be interpreted in terms of the energy cutoff in the fluctuation spectrum [20, 21]. This high temperature region is not in the focus of the present report.

The extraction of the coherence length was based on the imaginary part of the ac fluctuation conductivity σ_2 which is due only to the superconducting fluctuations, i. e. it has no contribution from the normal electrons. We may now check the consistency of the analysis on the real part of the ac fluctuation conductivity. By inserting the values of $\xi(T)/\xi_0$ from Fig. 2(b) in the real part of the solution of Eq. (2), we can calculate the corresponding values of σ_1. The result is plotted as the full line in Fig. 2(a). One observes an excellent agreement between the calculated and the total measured real conductivity up to $\epsilon = 0.09$. Beyond this region, the calculated values of σ_1 fall below the total experimental σ_1^{tot}. It appears that for $\epsilon < 0.09$, the normal conductivity contribution vanishes due to the decrease of the one-electron density of states at the Fermi level [22, 23]. However, for $\epsilon > 0.09$, there appears gradually a contribution of the normal conductivity which has to be added to the fluctuation conductivity σ_1 in order to get σ_1^{tot}. A more detailed analysis of this effect will be reported elsewhere [24].

We have also analyzed the same experimental data of the BSCCO-2212 sample using the expression for the ac fluctuation conductivity in the $2D$ case [16, 17]. Following the procedure analogous to the $3D$ case described above, we find the values of $\xi(T)/\xi_0$ for the $2D$ case which are also shown in Fig. 2(b). One finds that this coherence length saturates when T_c is approached. Since this is a completely unphysical behavior, one may conclude that BSCCO-2212 is not an effective $2D$ system near T_c. However, the analysis at higher temperatures reveals that both, $2D$ and $3D$ calculations yield nearly the same slope of $2/3$. The effective layer thickness in the $2D$ expression for the fluctuation conductivity is a free parameter which affects strongly the absolute values but only slightly the slopes of $\xi(T)/\xi_0$. In Fig. 2(b) we have used the effective layer thickness of 2.8 nm, which yields the best fit to the σ_1 data at higher temperatures, and in addition matches the coherence length of the $3D$ case. This remarkable result shows that the anisotropic $3D$ behavior matches with the $2D$ behavior when the coherence length $\xi_c(T)$ approaches the cutoff value ξ_{0c}/Λ_c.

One can check also the calculated σ_1 for the $2D$ case. The dashed line in Fig. 2(a) shows a strong deviation from the experimental data at temperatures close to T_c. This proves again that the behavior in this temperature range is not of the $2D$ nature. At higher temperatures, however, the calculated σ_1 in the $2D$ and $3D$ cases practically overlap, thus corroborating the result found in the coherence length.

Fig. 3 shows the final results of the analysis of the fluctuation conductivity in YBCO, and BSCCO-2223 films. From the temperature dependences of the reduced coherence lengths one concludes readily that the appearance of multiple critical regimes of the superconducting fluctuations is a general feature in high-T_c layered superconductors. The anisotropy of the system affects the temperature ranges of the critical regimes. The more anisotropic superconductors have their critical regimes extended to higher temperatures. BSCCO-2223 exhibits the critical state with $\nu = 1$ up to 1.4 K above T_c, which is considerably more extended than found above in BSCCO-2212. The $3D$ XY critical regime with $\nu = 2/3$ is found in BSCCO-2223 in a large interval $0.08 < \epsilon < 0.44$.

YBCO is the least anisotropic material of the three superconductors studied here. We observe in Fig. 3 that it also develops the critical regime with $\nu = 1$, but very close to T_c. The values of the reduced coherence length $\xi(T)/\xi_0$ are
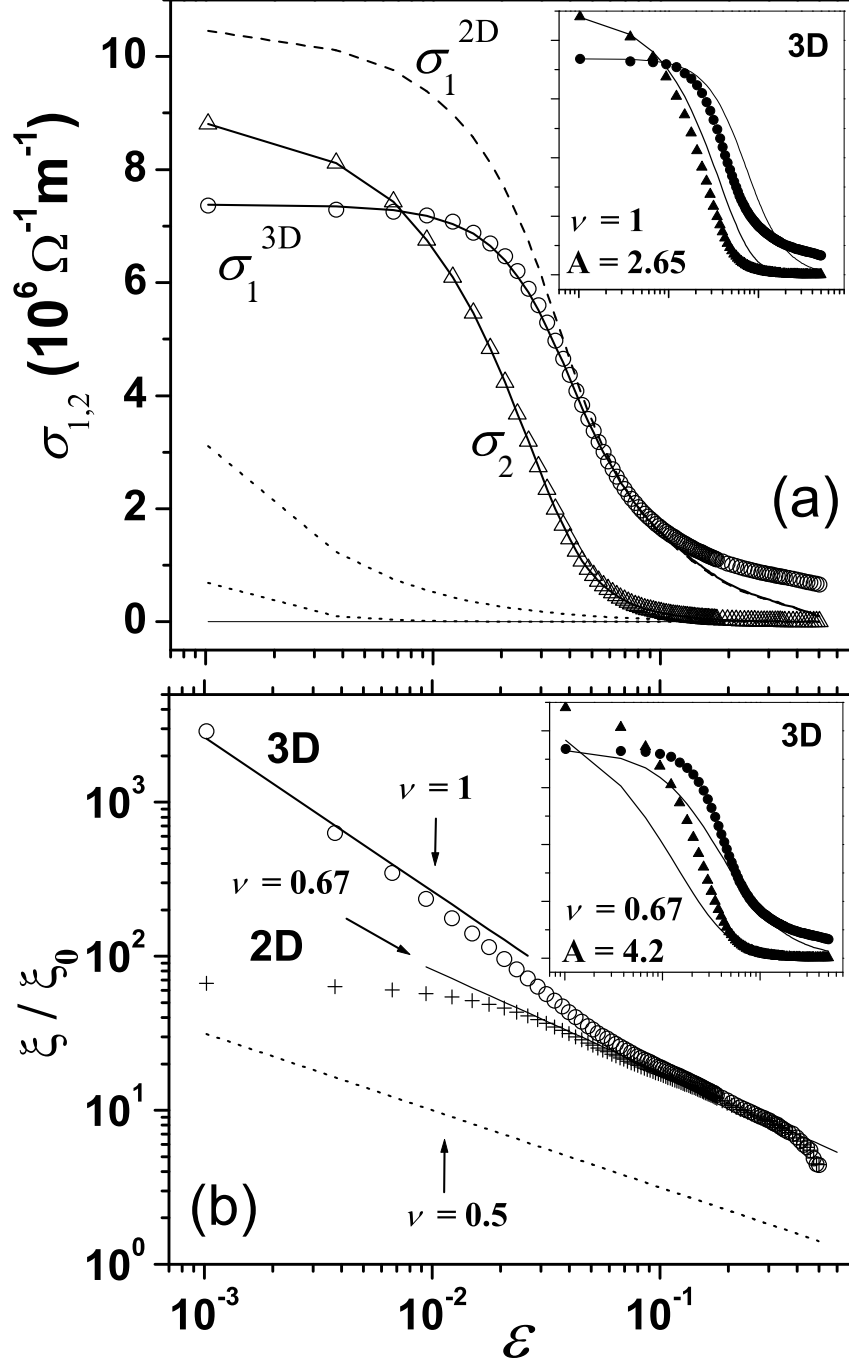
FIG. 2: (a) Temperature dependences of the total measured real ($\bigcirc$) and imaginary ($\triangle$) part of the complex conductivity above T_c in a BSCCO-2212 thin film. The dotted lines are the conductivity calculated using the Gaussian form of the coherence length. The full and dashed lines are the results of the selfconsistent *3D* and *2D* calculations, respectively, as explained in the text. (b) The reduced coherence length deduced from the experimental data of σ_2 by means of the *3D* ($\bigcirc$) and *2D* (+) theoretical expressions. The full lines show the slopes $\nu = 1$ and $\nu = 2/3$ for the two critical regimes. The Gaussian form $1/\sqrt{\epsilon}$ is shown by the dotted lines. The insets show the experimental data compared with the *3D* calculations using the coherence lengths of the form $(\xi(T)/\xi_0) = A/\epsilon^{\nu}$.
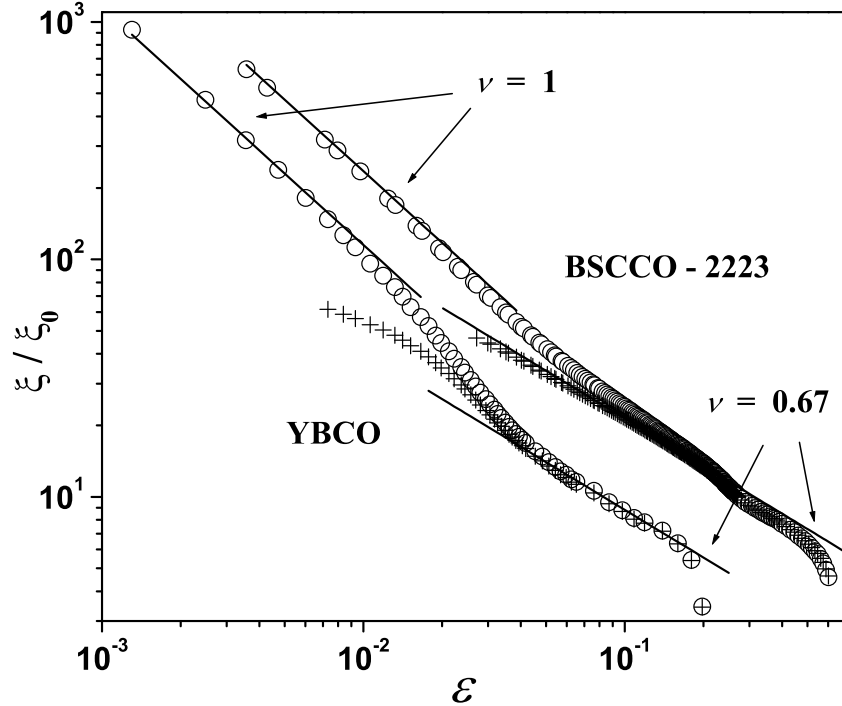
FIG. 3: Reduced coherence lengths in the high-T_c superconductors YBCO and BSCCO-2223 with various degrees of anisotropy. Both, 3D($\bigcirc$) and 2D($+$) results are presented.

much lower than in the BSCCO superconductors. Also, the *3D* XY critical regime with $\nu = 2/3$ is found in YBCO in a much narrower temperature range $0.05 < \epsilon < 0.12$. This finding is in agreement with the combined analysis of the *dc* conductivity, specific heat, and susceptibility measurements in YBCO single crystals [25]. Beyond $\epsilon = 0.12$, the reduced coherence length in YBCO shows a drop towards the Gaussian value $1/\sqrt{\epsilon}$. With such low values of $\xi(T)/\xi_0$ the *ac* fluctuation conductivity becomes so small that our signal to noise ratio precludes a further analysis.

In conclusion, we have determined for the first time the absolute values and the temperature dependence of the reduced coherence length $\xi(T)/\xi_0$ above T_c in high-T_c superconductors. Our results show in detail how the fluctuations of the order parameter develop in YBCO, BSCCO-2212, and BSCCO-2223 systems with different degrees of anisotropy. In the vicinity of T_c, one finds a hitherto unobserved critical regime with the static critical exponent $\nu = 1$. At higher temperatures one observes a crossover to a critical regime having $\nu = 2/3$ as predicted for the *3D* XY universality class.

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